Transport Times and Anthropogenic Carbon in the Subpolar North Atlantic Ocean.

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Abstract

Simultaneous measurements of chlorofluorocarbons, tritium, and helium-3 are used to estimate the distribution of surface-to-interior transit times within the subpolar North Atlantic ocean. The observed relationships among the different tracers implies that the transit-time distributions are broad, indicating that mixing plays an important role in transport over decadal timescales. Broad transit-time distributions are further shown to be consistent with the observed time variations of mid-depth tritium in Newfoundland Basin and Northeastern Atlantic between 1972 and 1997. We use the transit-time distributions inferred from the tracers to estimate the distribution, and change over the last two decades, of anthropogenic carbon in the subpolar North Atlantic. The values obtained are smaller than previous estimates using methods that have assumed weak mixing, with largest differences occurring in the Newfoundland Basin and Northeastern Atlantic.

Keywords: Tracers, Transit Time Distribution, Anthropogenic Carbon, Labrador Sea Water, North Atlantic Ocean.

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1 Introduction

Deep convection and formation of large volumes of nearly homogeneous water occur within the subpolar North Atlantic ocean, and these waters spread out into the North Atlantic and beyond (e.g., Talley and McCartney 1982), carrying high concentrations of dissolved oxygen, chlorofluorocarbons (CFCs), and anthropogenic carbon. Quantifying the timescales and pathways of this spreading is important for not only understanding ocean circulation but also regional and global climate.

Information on the spreading of these waters has been obtained from a variety of measurements, including those of salinity and potential vorticity (e.g., Talley and McCartney 1982), chemical tracers with time-varying sources or sinks (so called "transient tracers") (e.g., Fine 1995, Schlosser et al. 2001, Rhein et al. 2002), anomalies in hydrographical tracers (e.g., Sy et al. 1997, Curry et al. 1998, Koltermann et al., 1999), and subsurface floats (Lavender et al. 2000, Fischer and Schott 2002). However discrepancies exist between timescales derived from different tracers. For example, estimates of the spreading time of Labrador Sea water (LSW) from the Labrador Sea to the Northeastern Atlantic vary from 4-5 yrs from temperature and salinity anomalies (Sy et al. 1997) to greater than 10 yrs from CFCs (e.g., Fine et al. 2002), while subsurface floats have been observed to make this transit in only 2 yrs (Fischer and Schott 2002).

We show that these differences are a natural consequence of mixing. Most studies have assumed weak mixing, so that a single timescale adequately summarises the transport. However, in advective-diffusive flows, such as the oceans, there is a not a single transit time but rather a distribution of transit times from one location to another (e.g., Beining and Roether 1996, Deleersnijder et al. 2001, Haine and Hall 2002, Khatiwala et al 2001). Because of this distribution of transit times different tracer signals propagate at different rates, and the times derived from tracers are generally not fundamental timescales of the transport (Waugh et al. 2003, Wunsch 2002). While this complicates the interpretation of tracer timescales, it also allows tracers of different time-dependencies to be used in combination to estimate transit-time distributions (TTDs) of a flow (Waugh et al. 2002, 2003).

We use measurements of halocarbons (CFC11, CFC12, CFC113, and CCl_4), tritium, and helium-3 from several World Ocean Circulation Experiment (WOCE) cruises, between 1988 and 1997, to constrain the surface-to-interior TTDs in the subpolar North Atlantic ocean. We examine the relationships among the different transient tracers, and show that the observed tracer-tracer relationships are inconsistent with weak mixing and imply that the TTDs are broad and that mixing plays an important role in transport over decadal timescales. Broad TTDs are further shown to be consistent with the observed time variations of LSW tritium in Newfoundland Basin and Northeastern Atlantic between 1972 and 1997.

Given estimates of the TTDs it is possible to construct the interior concentration of conserved tracers from the tracer's surface time series. An important application of this approach is estimating the distribution of anthropogenic carbon in the oceans (Thomas et al. 2001, Hall et al. 2002). In these calculations, the surface history of anthropogenic dissolved inorganic carbon (ΔDIC) is reconstructed from the history of atmospheric CO_2 and the equilibrium inorganic carbon system, and is then propagated into the ocean interior using the TTDs. We apply this method to the WOCE data to estimate the distribution of ΔDIC in the subpolar North Atlantic, as well as the change in concentration over the last

two decades. We compare these to previous estimates that either (1) propagate the surface history into the interior using CFC ages as the water mass age (Thomas and Ittekot 2000, McNeil et al. 2003), or (2) uses Redfield ratios to account for biochemical sources and sinks (Gruber 1998, Wanninkhof et al. 1999, Koertzinger et al. 1998, 1999, Gruber and Sarmiento 2002). These previous methods all assume weak mixing. Our ΔDIC estimates using TTDs are in most instances smaller than previous estimates, with largest differences occurring in the Newfoundland Basin and Northeastern Atlantic.

2 Method

The approach used to constrain the transit time distributions (TTDs) is similar to that of Waugh et al. (2002, 2003). Measurements of transient tracers with different time dependencies are used to determine the range of possible values of the first two moments of the TTDs. The basis of the approach is the fact that, for steady transport, the interior concentration $c(r,t)$ of a tracer with spatially uniform, time-varying surface concentration $c_0(t)$ is given by (Hall and Plumb 1994, Haine and Hall 2002)

$$c(r,t) = \int_0^\infty c_0(t-t')\mathrm{e}^{-\lambda t'}\mathcal{G}(r,t')dt', \tag{1}$$

where $\mathcal{G}(r,t)$ is the TTD at location r and the $\mathrm{e}^{-\lambda t'}$ term is for the radioactive decay of tracers with decay rate λ. Given interior concentration and surface time series of a particular tracer this relationship can be inverted to place constraints on $\mathcal{G}$.

We assume, as in our previous studies, that the TTDs are Inverse Gaussian functions (Chhikara and Folks 1989, Seshadri 1999), i.e.,

$$\mathcal{G}(t) = \sqrt{\frac{\Gamma^3}{4\pi\Delta^2 t^3}}\exp\left(\frac{-\Gamma(t-\Gamma)^2}{4\Delta^2 t}\right), \tag{2}$$

where

$$\Gamma = \int_0^\infty \xi\mathcal{G}(\xi)d\xi$$

is the mean transit time ("mean age") and

$$\Delta^2 = \frac{1}{2}\int_0^\infty (\xi-\Gamma)^2\mathcal{G}(\xi)d\xi$$

defines the width Δ of the TTD. A given interior concentration of a transient tracer then limits Γ and Δ, for that location, to a range of values. Measurement of the concentration of a second tracer with sufficiently different time variations can further limit this range of values, and in some circumstances Γ and Δ can be tightly constrained.

In the analysis below we calculate the relationship between different tracers for families of TTDs with the same Δ/Γ ratio. The ratio Δ/Γ is a measure of the breadth of the TTD and importance of mixing, with Δ/Γ=0 corresponding to purely advective flow. For one-dimensional flows with constant velocity u and diffusivity k the TTDs are of the form (2), and the Peclet number of the flow $Pe = ux/k$ equals $(\Gamma/\Delta)^2$ (e.g., Waugh and Hall 2002). Therefore, a larger Δ/Γ can be interpreted as a smaller Peclet number.

To calculate tracer concentrations at interior locations using equation (1) it is necessary to know the surface time series of the tracers. For the halocarbons (CFC, CCl_4) we use the atmospheric time series of Walker et al. (2000), solubility functions from Warner and Weiss (1985), Bu and Warner (1995), and Hunter-Smith et al.(1983), and assume that the surface waters are at 90% saturation. The tritium surface time series is based on that of Dreisigacker and Roether (1978) and Doney and Jenkins (1994), and has been extended to 1997 using the WOCE surface measurements.

In our analysis we convert CFC concentrations into pCFC ages, $\tau_{\rm CFC}$, defined as the elapsed time since the surface concentration was equal to the interior concentration. This conversion accounts for variations in the CFC distributions due to variations in the temperature and salinity. Although it is possible to calculate an age using tritium and excess helium (the component of helium-3 that has come from tritium decay) together (e.g., Jenkins and Clarke 1976), we analyse each of these tracers separately.

3 Data

We use measurements of halocarbons (CFC11, CFC12, CFC113, and CCl_4), tritium, and helium-3 from World Ocean Circulation Experiment (WOCE) lines A1, A2, and A16 (see Figure 1). These lines pass through the Labrador and Irminger Seas, the Iceland, west European, and Newfoundland Basins, and the Rockall Trough, and the measurements sample a range of water masses including subpolar mode water, Labrador Sea water (LSW), and Demark Strait and Iceland overflow waters.

Measurements of CFC11, CFC12, tritium and helium-3 were made along A1 in 1991 and 1994, along A2 in 1994 and 1997, and along A16 in 1988. CFC113 and CCl_4 were also measured on the two A2 cruises. The CFC, tritium, and helium-3 data (and the relationships among these tracers) from A16 has been examined by Doney et al. (1997), while Fleischmann et al. (2001) examined the CFC11 and tritium data from both cruises along A1 and the 1994 A2 cruise.

We also consider earlier measurements of tritium in the North Atlantic subpolar ocean. Tritium measurements were made in the western part of the subpolar gyre in 1972 as part of the Geochemical Ocean Sections Study (GEOSECS) (Ostlund and Rooth 1990). Repeat measurements at some of these sections and also in the eastern part of the gyre were made in 1981 as part of the Transient Tracers in the Ocean (TTO) study (Ostlund and Rooth 1990). Measurements were also made in the subpolar North Atlantic in 1975 as part of a Charlie-Gibbs Fracture Zone (CGFZ) study (Top et al. 1987) and in 1986 as part of the TOPOGULF study (Andrie et al. 1988). We focus here on measurements made in the Newfoundland and west European Basins, near the WOCE A2 or A16 lines. In particular, GESOSECS station 27, CGFZ station 702, TTO stations 117, 118, and 228, and TOPOGULF stations 83 and 110, see Figure 1.

4 Transit-Time Distributions

We first consider the relationships between tritium and CFC12 measurements. The symbols in Figure 2 show the observed relationship between tritium and CFC12 age ($\tau_{\rm CFC12}$) for each

of the five WOCE cruises. The relationships are remarkably similar among cruises, with the only significant difference being lower tritium for later years, consistent with the decay of tritium to helium. All data from the cruises are shown, but the relationships are still very compact: There is little dependence on basin (e.g., the 1994 A1 cruise samples the Labrador and Irminger seas as well as the Iceland Basin and the Rockall Trough) and a smooth transition with depth.

The curves in Figure 2 show the modeled tracer relationships for TTDs with different Δ/Γ ratios. The two-parameter TTD of form (2) is completely specified by two independent constraints, in this case τ_{CFC12} and a choice of the ratio Δ/Γ. (Each point on a curve in Figure 2 corresponds to a particular value of Γ and Δ, with both values increasing for increasing τ_{CFC12}.) The modeled relationships for small Δ/Γ, which correspond to narrow TTD and advectively dominated flow, are inconsistent with the observations for water masses with $\tau_{\mathrm{CFC12}} > 10$ yrs. For narrow TTD a large, well-defined, peak in the tritium is predicted for τ_{CFC12} between 20 and 30 yrs (the time since the mid-1960s peak in the surface tritium), but such a peak is not observed. There is, however, good model-data agreement for TTDs with $\Delta/\Gamma \approx 1$, with the model curves reproducing the observed variation of tritium with τ_{CFC12}, for all values of τ_{CFC12}. These large values of Δ/Γ correspond to broad TTDs, and hence a large range of transit times.

Although there is good model-data agreement for $\Delta/\Gamma = 1$ for all cruises, the value of Δ/Γ that best fits the observations varies among the different WOCE lines: The best fit Δ/Γ is 0.75 for A16, 1.05 for A2, and 1.3 for A1. It is unclear whether these differences are due to real differences in the transport or due to uncertainties in the measurements and the assumed tracer surface time series. The surface history of tritium is uncertain, especially during the 1960s and 1970s, and although the CFC12 surface history is well known the degree of saturation in surface waves is uncertain. In the calculations presented we have assumed a saturation of 90% for CFC12 and used a tritum surface history based on that of Doney and Jenkins (1994). We have repeated the above analysis varying the CFC saturation and tritium time series by 10%, and the resulting variations in the best fit Δ/Γ is about 20%, similar to the variation between WOCE lines. Because of these uncertainties (and also the assumption of a particular form of the TTDs, see discussion below) it is not possible to determine a precise value of Δ/Γ. However, the general conclusion that the TTDs are broad and there is a wide range of transit times still holds.

The relationships among the other transient tracers measured also place constraints on Δ/Γ, and can be used to test the above inferences from tritium and CFC12. Figure 3 shows the relationships of the other transient tracers with τ_{CFC12} for measurements made along A2 in 1997 (symbols) and model TTD calculations (curves). (Very similar relationships hold for the other cruises.)

The ages from the measurements of CFC11, CFC113, and CCl_4 are, in general, different from those from CFC12. This is expected for flows with mixing because of the different temporal histories of the tracers (Waugh et al. 2003). Furthermore, for all the halocarbons, the observed relationships for waters colder than 5°C are inconsistent with $\Delta/\Gamma \approx 0$ and indicate that $\Delta/\Gamma \approx 1$. Hence, as with the tritium-τ_{CFC12} relationships, the relationships among the halocarbon ages indicate that the TTDs are broad and there is strong mixing.

The observed relationships of τ_{CFC113} and τ_{CCl4} with τ_{CFC12} for warmer waters ($T > 5$°C) are not consistent with model calculations even for very large Δ/Γ. The observed τ_{CFC113}

and τ_{CCl4} are higher than modeled. These differences are consistent with the biochemical degradation of these tracers in warm waters (e.g., Roether et al. 2001 Wallace et al. 1994, Huhn et al. 2001). Biochemical loss results in lower concentrations which, if not accounted for, appear as older ages. However, even if only cold waters are considered the modeled τ_{CCl4} for Δ/Γ larger than 1 is less than observed. This could be because of CCl_4 loss in cold waters, at a slower rate than in warmer waters. If we assume that the loss is a first-order process with loss rate λ (i.e. the loss equals a constant λ times the concentration) we can obtain an estimate of λ by using the TTDs implied by other tracers (e.g., tritium and CFC12) in equation (1) and determining the λ that reproduces the observed τ_{CCl4}:τ_{CFC12} relationships. Figure 5 shows the observed τ_{CCl4}:τ_{CFC12} relationships (for $T < 5^oC$) from the two A2 crusies together with model curves for TTDs with $\Delta = \Gamma$ for varying λ. For both data sets there is some scatter and λ can only be constrained to a range of values, i.e., the observations generally fall within curves for $\lambda = 0.005$ and 0.015 yr^{-1}. Repeating these calculations assuming TTDs with Δ/Γ equal to 0.75 or 1.25 produces very similar results. This loss rate is higher than the values of less than 0.1% per year determined by Huhn et al. (2001) (from measurements in the South Atlantic), but still represents very slow loss. Repeating this analysis for slightly warmer waters yields larger loss rates: For temperatures between 5 and 7.5°C the loss is around 3% per year, while for temperatures between 7.5 and 10°C the loss is around 8% per year (not shown). Hence, as in Huhn et al. (2001), our analysis indicates that the loss rate increases very rapidly with temperature.

The observed relationship between excess helium and τ_{CFC12} also implies strong mixing ($\Delta \approx \Gamma$) (Figure 3d). However, in contrast to the other tracer-tracer relationships, the modeled relationship between excess helium and τ_{CFC12} is sensitive to Δ/Γ for large values. This suggests that the observed relationship may constrain the TTD more tightly than the other tracer pairs. Unfortunately there is a lot of scatter in the data, and no single Δ/Γ curve matches the data, with the data below the Δ/Γ =1 curve for young τ_{CFC12} and above for old τ_{CFC12}. Lower excess helium in young water masses may be due to rapid loss of helium within the mixed layer to the atmosphere, whereas high excess helium in old water masses is likely due to primordial helium, which is not accounted for when calculating excess helium. The cause of the increased scatter for intermediate ages is unclear, and may be due uncertainties in the calculation of excess helium.

The excess helium - tritium relationship (not shown) has a similar "hook" shape as the above excess helium - τ_{CFC12} relationship, and is also reasonably well modeled by TTDs with $\Delta \approx \Gamma$. Similar excess helium - tritium distributions have been reported by Jenkins (1988) for TTO data, Andrie et al. (1988) for their 1983 data and Doney et al. (1997) for the A16 data. Jenkins (1988) noted that peak tritium values were much lower than expected for advective flow, and further showed that a one-dimensional model with Peclet number (Pe) around 1 best matches the TTO data. This is consistent with our above analysis as Δ/Γ = 1 corresponds to Pe = 1, for one-dimensional flow with uniform advection and diffusion.

As a final check on the above estimates of Γ and Δ we now examine the temporal variation of tritium. Tritium measurements have been made in the Newfoundland and west European basins several times since 1972 (see Section 3 and Figure 1), and these can be used to test whether the temporal variation is consistent with broad TTDs. Although the location of these measurements varies (see Figure 1) the measurements of LSW along A2 and A16 suggest that these locations have similar tritium and CFC-12 concentrations in LSW (i.e.

there are only small variations in LSW tracer concentrations along A2 between 45W and 20W and along A16 between 40N and 50N). The symbols in Figure 4 show the evolution of tritium at 1500 m for the measurements between 1972 and 1997, at the locations shown in Figure 1. The values shown from 1988 on are based on several profiles for each year, all of which show weak vertical gradients between 1000 and 2000 m. Because of the weak vertical gradients there is only weak sensitivity to the depth used to determine the tritium values. However, the 1970s values are based on single profiles for each year and these show large vertical gradients around 1500 m, so there is larger sensitivity to depth used to determine the tritium value.

Also shown in Figure 4 is the predicted evolution of tritium for TTDs with various Δ/Γ. For each value of Δ/Γ the value of Γ and Δ are chosen so that the tritium in 1994 is the same. Comparison with the observations shows that the observations are inconsistent with narrow TTDs ($\Delta/\Gamma \leq 0.5$), in agreement with the above analysis of tracer-tracer relationships. We draw the same conclusion even if we only use observations during and after 1988, for which there are numerous of measurements in LSW for each year and well defined tritium values. The best match with the observations occurs for $\Delta/\Gamma = 0.75$, again consistent with the tracer-tracer analysis. While the values from the 1970s would appear to limit Δ/Γ tightly, the constraint they impose is unfortunately weakened by the dearth of measurements and large vertical gradients. Nonetheless, the tritium data overall eliminate not only narrow TTD, but also indicate that $\Delta/\Gamma > 1.5$ is unrealistic.

The above analysis shows that Inverse Gaussian TTDs with $\Delta \approx \Gamma$ can reproduce the observed tracer-tracer relationships among several transient tracers for cruises from several years as well as the observed temporal variations of tritium in LSW between 1972 and 1997. As mentioned above, such TTDs are very broad and correspond to a large range of transit times. This is illustrated in Figure 6(a) where the TTDs with $\Delta = \Gamma$ and τ_{CFC12} (in 1994) = 15, 20, and 25 yrs are shown. These TTD are asymmetric, have narrow peaks at transit times much younger than the mean transit time Γ (symbols), and have long tails representing old water components. Importantly, Γ is much larger then the CFC12 ages. The difference between Γ and τ_{CFC12} for TTDs with $\Delta \approx \Gamma$ is shown in Figure 6(b). There are large differences between the two "ages" for CFC12 ages greater then 15 yrs. This shows that for much of the subpolar gyre CFC12 ages are much younger then the mean transit time.

A source of uncertainty in our analysis arises from the use of a particular functional form for the TTD, in this case the Inverse Gaussian form. Waugh et al. (2002; 2003) examined the sensitivity to the shape of the TTD, looking at the impact of bimodality on tracers. They found that, for tracers whose history spans the TTD, once the first and second moments (Γ and Δ) are determined, there is little additional sensitivity to higher moments. In fact, to the extent a tracer's surface history can be modeled as a quadratic function in time, only the first two moments of the TTD effect its concentration (Waugh et al., 2003). On the other hand, the magnitude of water components at transit times older than the history of the tracers cannot be constrained by the tracers. Thus, for example, one cannot rule out, on the basis of the above transient tracers alone, a second peak in the TTD at transit times beyond 40 to 50 years. (Because of its longer atmospheric history τ_{CCl4} may provide some information, but its slow oceanic loss complications interpretation of the measurements.)

5 Anthropogenic Carbon

An important application of transient tracers is the estimation of anthropogenic carbon distribution and uptake. The fact that there is a wide range of transit times due to mixing needs to be considered in this application. Several different methods to infer anthropogenic carbon have been developed, and there are considerable discrepancies among the estimates (e.g., Wallace et al., 2001; Wanninkhof et al., 1999). Most methods assume mixing to be weak, which, in light of our tracer analysis, appears to be erroneous in the subpolar North Atlantic. A new method has been proposed by Hall et al. (2002) that does not assume mixing to be weak, and, in addition, does not require the use of uncertain Redfield ratios to account for biochemical sources and sinks of carbon (see also Thomas et al., 2001). In this method, TTDs inferred from tracers are used to propagate to the interior an estimate of the surface history of ΔDIC. As in most other studies, the surface ΔDIC history is calculated from equilibrium inorganic carbon chemistry, by assuming the air-sea CO_2 disequilibrium to be constant.

We calculate ΔDIC along A1 and A2 using the TTDs inferred from the CFC12 measured in 1994 assuming a fixed Δ/Γ for all data. Figure 7 shows the average vertical profiles ΔDIC, in 1993, for (a) Irminger Sea, (b) Iceland Basin, (c) Newfoundland Basin, and (d) West European Basin, for several different Δ/Γ. (As we assume steady transport we can calculate ΔDIC for any year, and values for 1993 are shown here to enable comparison with other estimates, see below.) There is only weak sensitivity of ΔDIC to the exact value of Δ/Γ for values within the range constrained by the transient tracers (i.e., $\Delta/\Gamma \approx 1$). However, for mid- and lower depths, there is a large difference between these values and those obtained assuming $\Delta = 0$, which corresponds to using the CFC age as the water mass age. The CFC12 history is shorter and more nonlinear than ΔDIC, and, therefore, CFC12 ages preferentially weight young components of broad TTDs, compared to ΔDIC. One then looks too recently in the ΔDIC surface history, causing overestimates in ΔDIC (Hall et al. 2002, Waugh et al. 2003). This overestimate increases with CFC age, and the difference between calculations of ΔDIC using different Δ/Γ is largest in the eastern part of the subpolar ocean where the ages are largest. For example, the column ΔDIC from $\Delta = 0$ calculations is 5, 30, 25, and 40% larger than from $\Delta = \Gamma$ calculations for the Irminger Sea, Iceland Basin, Newfoundland Basin, and west European Basins, respectively.

The distribution of ΔDIC shown in Figure 7 is qualitatively similar to previous estimates (e.g., Kortzinger et al. 1998, Thomas and Ittekot 2000). ΔDIC has penetrated throughout the water column in all basins, and the concentration decreases with depth and from west to east consistent with the increase in "age" of the water masses. An exception to the increase with depth occurs in the Irminger Sea where there is an increase at the bottom due to the presence of "young" Demark Strait overflow water.

Although there is qualitatively agreement with previous estimates there are significant quantitative differences. For example, the horizontal lines in Figure 7d show the ranges from several contemporaneous ΔDIC estimates for the same location and time (Wanninkhof et al. 1999, Koertzinger et al. 1998, 1999, Gruber and Sarmiento 2002), which use either the Gruber et al (1996) or Chen and Millero (1979) methods. Our estimates are lower than previous estimates in upper and middle levels, and within the range of other estimates for deep waters. Comparisons of our calculations with estimates of 1982 ΔDIC concentrations by

Gruber (1998), in both the eastern and western basins of the subpolar gyre, show differences consistent with these biases: Our estimates are lower in upper waters but similar or larger in deeper waters. The cause of the differences among the previous estimates is not clear. It is important to note even studies that ostensibly apply the same inference technique to the same data obtain different ΔDIC estimates. Nonetheless, some of the differences between our estimates and those from calculations using the Gruber method can be explained by biases in the treatment of transport. In upper and middle ("fully contaminated") isopycnals the use of tracer ages as the water mass age causes a high bias in ΔDIC. On deep isopycnals small, non-zero concentrations of ΔDIC on isopycnals believed to "uncontaminated" cause a low bias (Hall et al. 2002).

Accounting for mixing is also important for calculations of the change in ΔDIC over a certain period. McNeil et al. (2003) have recently calculated the ΔDIC uptake between 1980 and 1999 using CFC ages as the water mass age, which is equivalent to our $\Delta = 0$ calculation. However, as shown in Figure 8 the values calculated using $\Delta = 0$ are significantly larger than calculations assuming realistic values of Δ/Γ. Assuming no mixing results in an overestimate of the column uptake of ΔDIC (compared to calculations using $\Delta/\Gamma = 1$) by 15% in the Irminger Sea, 25% in the Iceland Basin, and greater than 30% in the Newfoundland and West European Basins. We conclude that the McNeil et al. (2003) "no mixing" calculations of DIC uptake in the subpolar North Atlantic ocean are a significant overestimate.

Hall et al. (2003) recently applied a modified, inventory-based TTD method to Indian Ocean CFC data, and also found a significant difference between weak and strong mixing calculations of the decadal-scale DIC uptake, with the weak-mixing limit about 30% larger than the strong-mixing limit.

Hall et al. (2003) also relaxed the constant-disequilibrium assumption made here, resulting in an additional reduction in ΔDIC of 6-9%. Their method to allow time-varying disequilibrium requires knowledge of ΔDIC over entire volumes bounded by isopycnals, and cannot be applied to the calculations here on specific WOCE lines. Allowing variable disequilibrium always acts to reduce ΔDIC estimates (Hall et al, 2003). Thus, our estimates, already lower than previous estimates, represent upper bounds on ΔDIC.

6 Concluding Remarks

The measurements of transient tracers in the subpolar North Atlantic ocean place tight constraints on the surface-to-interior transit time distributions (TTDs). The relationships among chlorofluorocarbons (CFCs), tritium, and helium-3 from five cruises (between 1988 and 1997) and the temporal variation of tritium in LSW between 1972 and 1997 all indicate that the TTDs are very broad. This implies that mixing plays an important role in transport the subpolar North Atlantic over decadal timescales, and must be taken into account when interpreting transient tracers.

One application of the tracer-inferred TTDs is the estimation of loss rates of non-conserved tracers. Here, we have estimated that the loss of CCl4 is about 1% per year for waters colder than 5°C, increasing rapidly with temperature. It may be possible to use a similar analysis to estimate biochemical loss rates of other, steady-state tracers, e.g., oxygen.

TTDs inferred from the transient tracer measurements can also be used to propagate

the surface history of anthropogenic carbon (ΔDIC) into the ocean interior, to estimate the distribution and uptake of ΔDIC. We have used this method to estimate ΔDIC in the subpolar North Atlantic, and obtained values that are, in most instances, smaller than previous estimates, which used methods that have assumed weak mixing. This is true even if only the uptake over the last two decades is considered, in which case use of CFC age as the water mass age overestimates the uptake by 20-30% in most of the subpolar North Atlantic.

Another application of TTDs is the interpretation of the propagation of temperature and salinity anomalies in LSW. The time lag in these anomalies has been used to estimate transit times from the Labrador Sea, and these transit times are generally much shorter than those inferred from CFCs (e.g., Sy et al. 1997), leading to some confusion. However, for flows with strong mixing the phase of high frequency variations propagate much faster than that of a slowly changing tracer (Hall and Plumb 1994, Waugh and Hall 2002), so given that there is strong mixing in the subpolar North Atlantic the propagation of LSW anomalies is expected to be much faster than that of CFCs. In fact, preliminary calculations using the TTDs determined in this study yield time lags in LSW anomalies that are much smaller than CFC lags and are broadly consistent with observations. We are currently performing a more detailed analysis of the propagation of LSW anomalies. This analysis may yield more refined constraints of the TTDs, including information on temporal variations in the transport.

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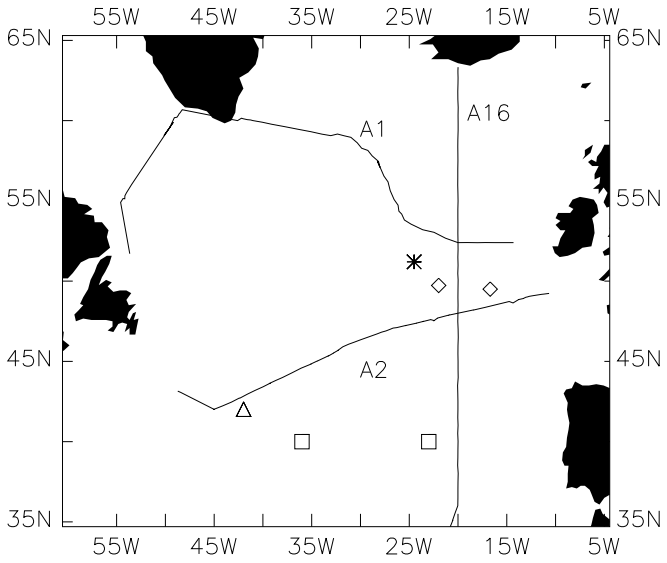

Figure 1: Map of WOCE cruise lines A1, A2, and A16. Symbols show sites of additional tritium measurements from GEOSECS (triangle), CGFZ (asterisk), TTO (triangle, diamonds), and TOPOGULF (squares) studies used in Figure 4.

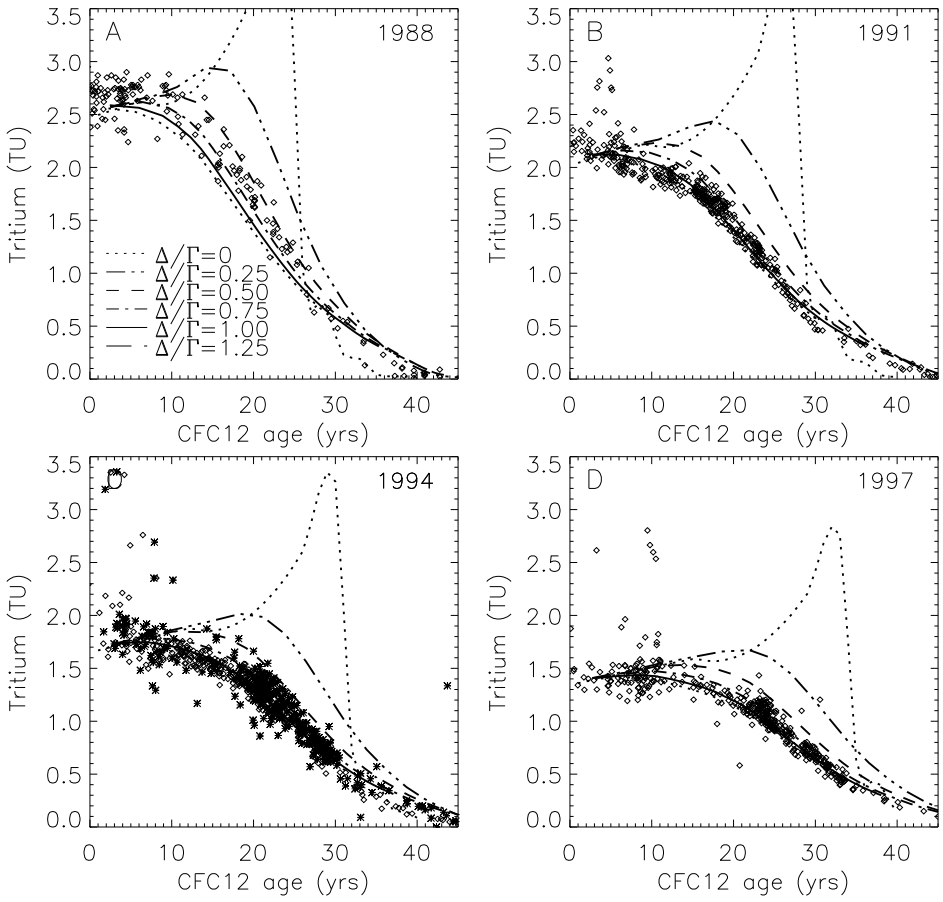

Figure 2: Tritium concentration (TU) plotted against τ_{CFC12} for data from WOCE cruises in (a) 1988 (A16), (b) 1991 (A1), (c) 1994 (plus symbols for A1 and diamonds for A2), and (b) 1997, together with predictions from model TTDs (curves) with different Δ/Γ ratios.

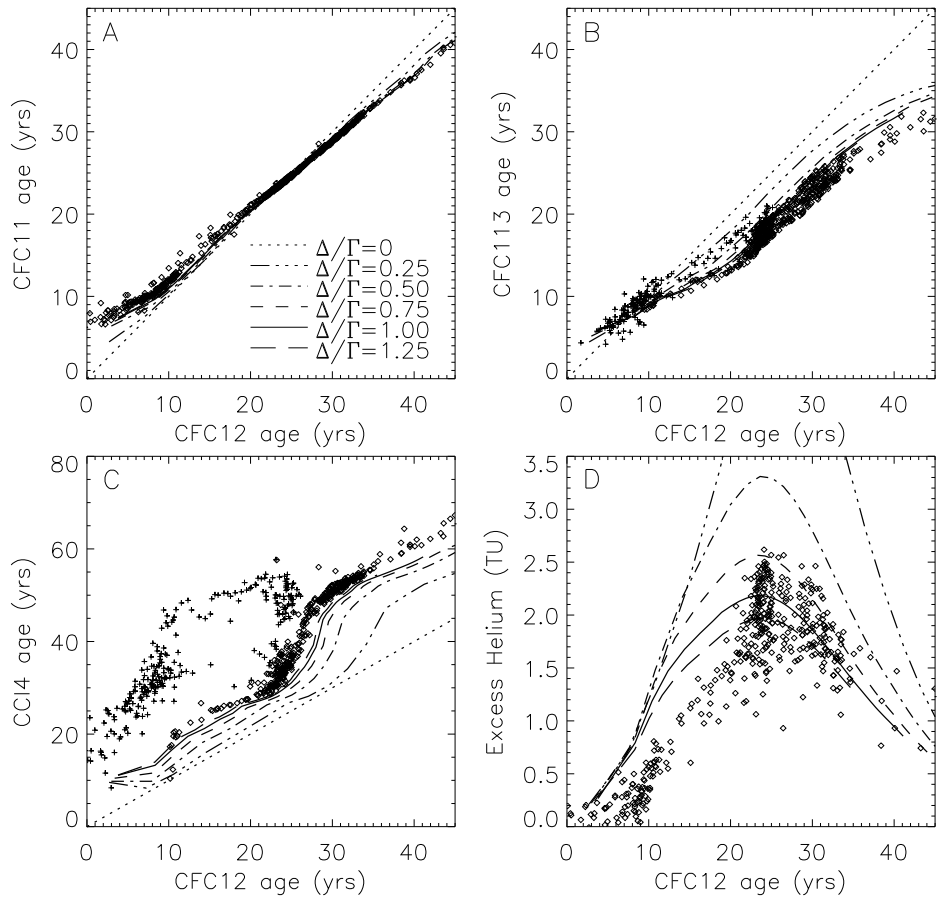

Figure 3: Relationships between (a) τ_{CFC11}, (b) τ_{CFC113}, (c) τ_{CCl4}, and (d) Excess Helium, with τ_{CFC12} for measurements along A2 in 1997 (symbols) and for model TTDs (curves). Plus symbols in panels (b) and (c) correspond to observations in water with temperatures warmer than 5°C.

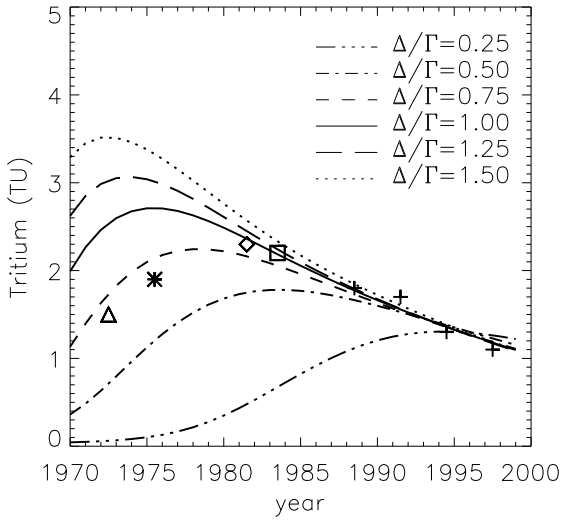

Figure 4: Observed time variation of tritium (symbols) at 1500 m in Newfoundland and West European Basins (see text and Figure 1) and predictions for TTDs with different Δ/Γ (curves). Values of Γ and Δ are chosen, for each Δ/Γ, so that tritium in 1994 is the same.

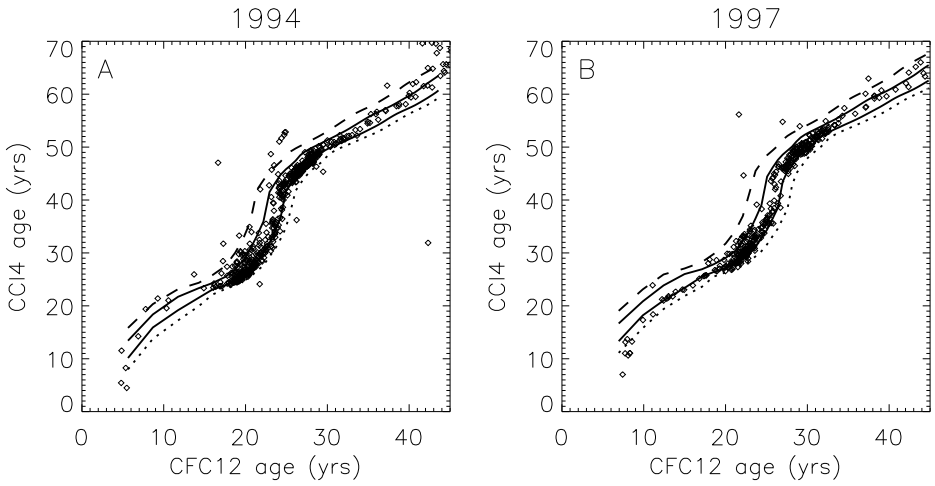

Figure 5: Relationships between τ_{CCl4} and τ_{CFC12} for measurements (in waters colder than 5°C) along line A2 (symbols) and for model TTDs (curves), for (a) 1994 and (b) 1997. The different curves correspond to λ equal to 0 (dotted), 0.005 yr^{-1} (lower solid), 0.015 yr^{-1} (upper solid), and 0.025 yr^{-1} (dotted).

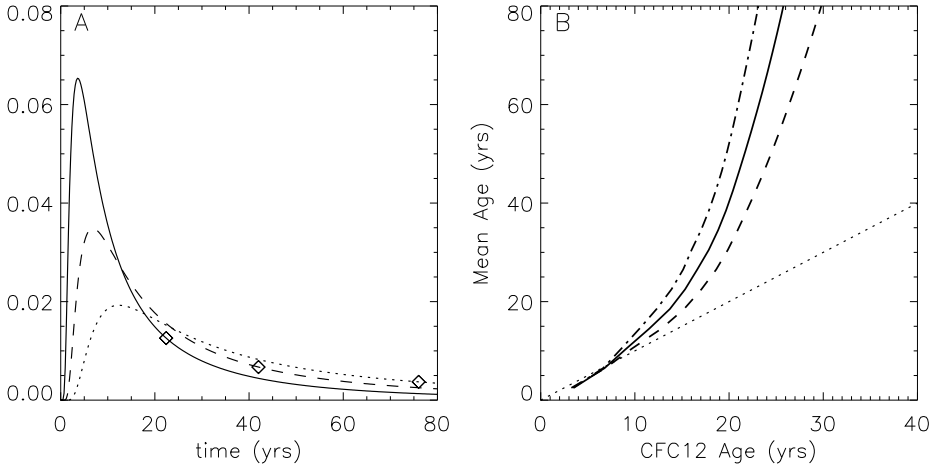

Figure 6: (a) TTDs with $\Delta = \Gamma$ that produce (in 1994) τ_{CFC12} equal to 15 (solid curve), 20 (dashed), and 25 (dotted) yrs. Symbols show Γ for each TTD. (b) Variation of Γ with τ_{CFC12} for TTDs with Δ/Γ equal to 0.75 (dashed), 1.0 (solid), and 1.25 (dotted).

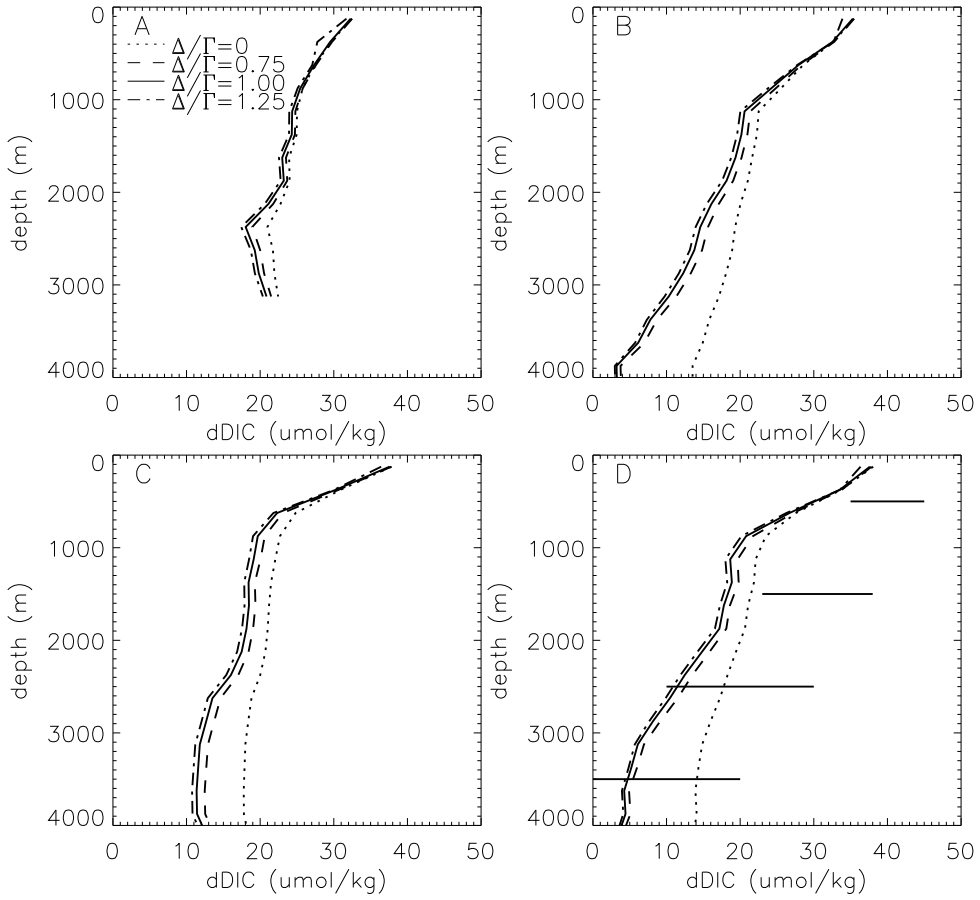

Figure 7: Average vertical variation of ΔDIC in 1993 for (a) Irminger Sea, (b) Iceland Basin, (c) Newfoundland Basin, (d) west European Basin, calculated from CFC12 data and TTDs with different values of Δ/Γ. Horizontal lines in (d) are the range of previous estimates for this region.

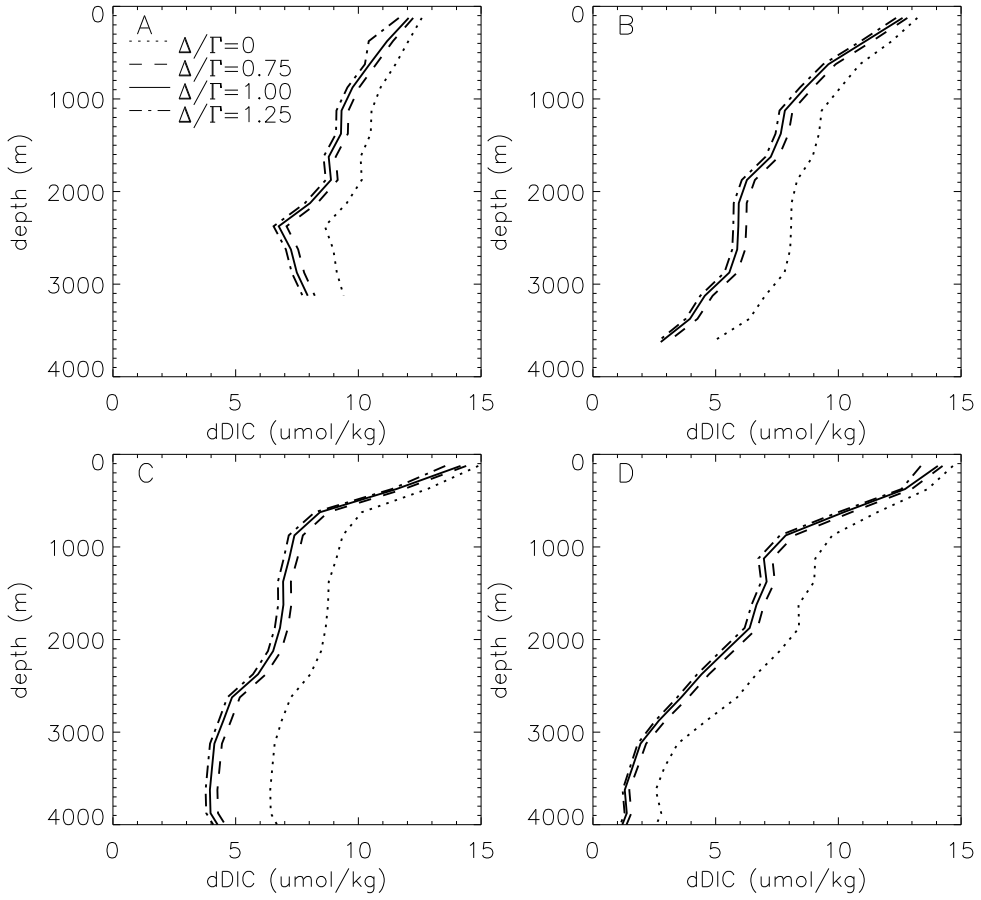

Figure 8: As in Figure 7 except for uptake of ΔDIC between 1980 and 1999.